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Shear Effects on Cholesteric Liquid Crystals

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Abstract—On shearing, cholesteric liquid crystal (L.C.) films lose color intensity, but the wavelength of their maximum reflectance does not change appreciably. The rate of recovery of the intensity after cessation of shearing is linear with the logarithm of time.

The thickness of the liquid crystal film is not a very strong factor in the behavior of total reflectance as a function of shear rate; however, the decrease in wavelength of maximum intensity as a function of shear is smallest for the thickest L.C. films.

Explanations for the large intensity drop combined with the almost stationary peak reflectance wavelengths, on shearing, are offered in terms of a mechanical model.

1. Introduction

Liquid crystals which have been de-aligned by exposure to heat or to electric fields can often be re-aligned by the application of a shearing motion to their surfaces. Some cholesteric liquid crystal systems, in fact, are more sensitive to shear than to heat or electric stimuli, and give well-defined and very bright color patterns.

Since color response in cholesteric liquid crystals is a function of their inter- and intra-molecular structures, it is reasonable to assume that shear-induced color patterns are also dependent upon the structure of the liquid crystals.

Previous studies of the effect of shear on liquid crystals have predominantly involved the use of high frequency sound waves. Using this technique, smectic,[1] nematic,[2] and cholesteric[3] compounds have been examined. The effect of mechanical shear has received little attention; one example is reported by Adams *et al.*,[3] who examined a binary cholesteric mixture before and after mechanical disturbance. We now wish to report the effect of continuous mechanical shear upon a cholesteric mixture.

2. Experimental

The apparatus used to perform the shear experiments is shown in Fig. 1. It consisted of an optically ground flat glass plate which was

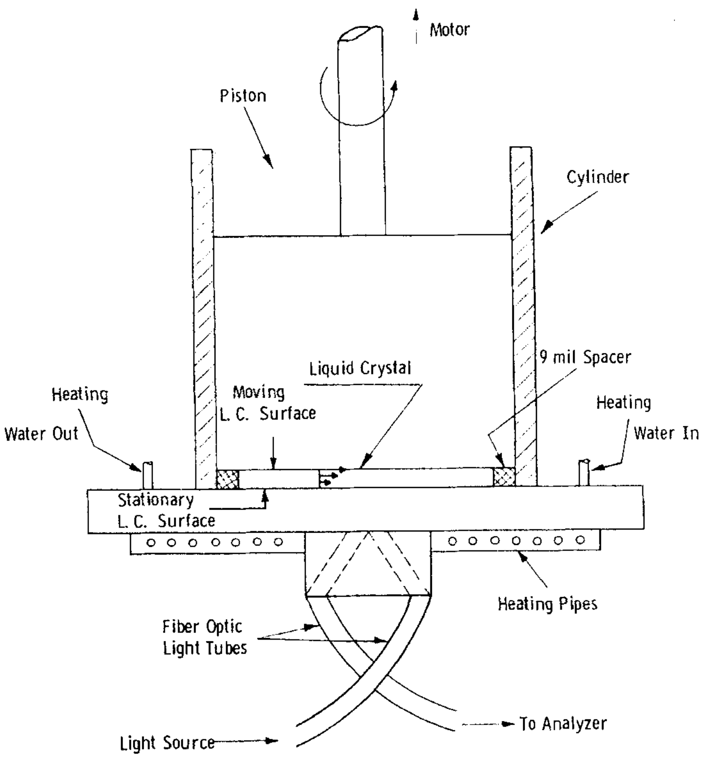

Figure 1. Apparatus for the study of shear effects on the color response of liquid crystals.

securely anchored to a heavy base. A Plexiglas cylinder was attached to the plate and a specially ground Plexiglas piston was fitted to the cylinder. The bottom of the piston was fitted with a glass plate which was ground to mate perfectly with the stationary plate. Spacers of appropriate size and thickness were attached to the bottom of the piston to obtain desired film thicknesses. The liquid crystal temperature was controlled by heating pipes from a recirculating water bath, attached to the bottom of the stationary plate. The apparatus was designed to fit on an available Cary Spectrophotometer. The light source from the spectrophotometer was conducted to a point on the bottom plate, 1 cm from the axis, by fiber optic light pipes (0.5 cm diam), and the reflected light was conducted to the analyzer in a similar manner. The piston was made to revolve by means of a low rpm, high torque motor.

The liquid crystal samples were composed of 23% cholesteryl nonanoate (CN), 44.5% oleyl cholesteryl carbonate (OCC), and 32.5% cholesteryl chloride (CC) by weight. This formulation was considered to be relatively temperature insensitive.

Experiments were carried out to determine: (1) the relation between the reflected wavelength of maximum intensity and the rate of shear; (2) the effects of temperature on the intensity and on the wavelength of maximum reflectance; (3) the relaxation time, or the time necessary for the sample to regain its original optical properties after cessation of shearing; (4) the effect of the rate of shear on the intensity at maximum reflectance; (5) the effect of film thickness on the total reflectance, the reflectance limits, the wavelength of maximum intensity, and on the intensity of maximum reflectance.

3. Results

Test samples (0.025 cm thick) at 24 °C exhibited a very sharp peak of reflected light at 590 nm and zero shear ($V = 0$). This is shown in Fig. 2; any shearing action reduced the intensity of reflected light at 590 nm with a small change in the position of the peak reflectance wavelength. A change in color observed during these experiments may have arisen from the fact that there is an increase in the amount of shorter (blue) wavelength light that is reflected while shearing takes place ($V = 1$ cm/min, Fig. 2).

Figure 3 shows that the reduction in peak height, as given by $I(\omega)/I(\omega = 0)$ (a measure of the intensity of the reflected light), was linear with the logarithm of the velocity of the rotating plate, over the experimental range. This velocity can be converted to shear rate using a relationship of the form: $\tau = KV^n$ where τ is shear rate, V is

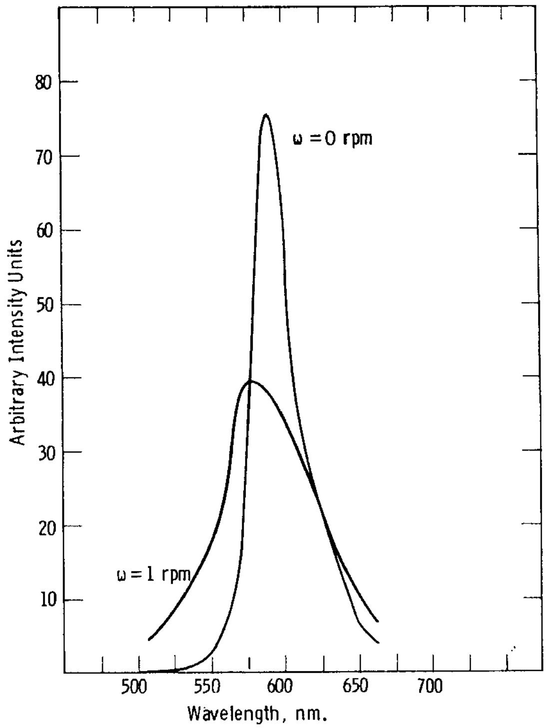

Figure 2. Color intensity peak under shear ($V = 1$ cm/min) and at rest ($V = 0$ cm/min) for a 0.025 cm film.

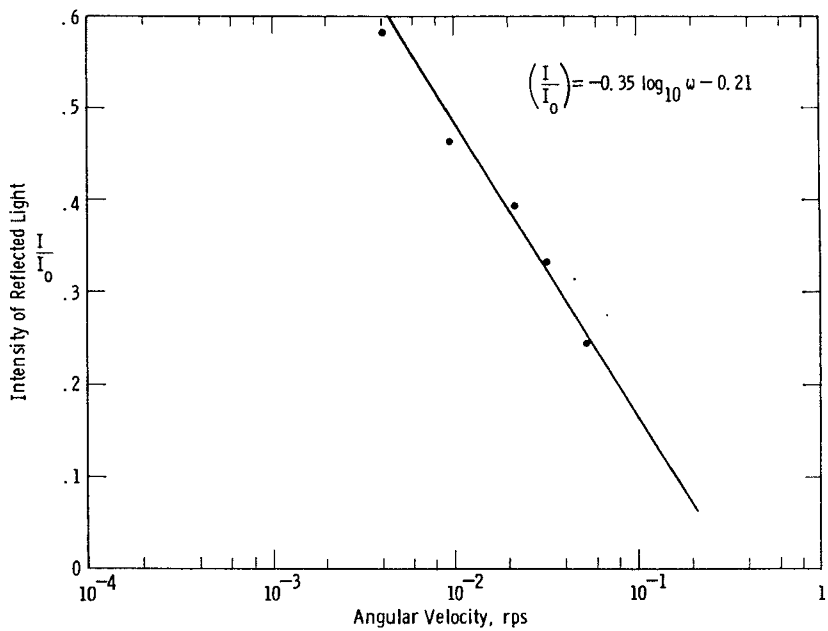

Figure 3. Decrease of peak height as a function of shearing for a 0.025 cm film.

the velocity of the piston, K, n, are constants for the particular material under consideration and the geometry used.

It was important to maintain the liquid crystal samples at constant temperature because the position of the peak wavelength was found to be strongly temperature dependent under shear. On the other hand, the peak intensity was relatively insensitive to temperature change.

An interesting phenomenon was noticed when a particular wavelength was examined as the shearing motor was turned off. The peak wavelength, instead of immediately regaining the intensity it exhibited prior to shearing, regained its original magnitude (I_0) in a fashion that was found to be linear with the logarithm of time. Figure 4 shows the relaxation time curves at different temperatures. It was expected that the rate of recovery would be strongly temperature dependent, but the slope of the curves at 40°, 34°, and 24 °C are essentially the same.

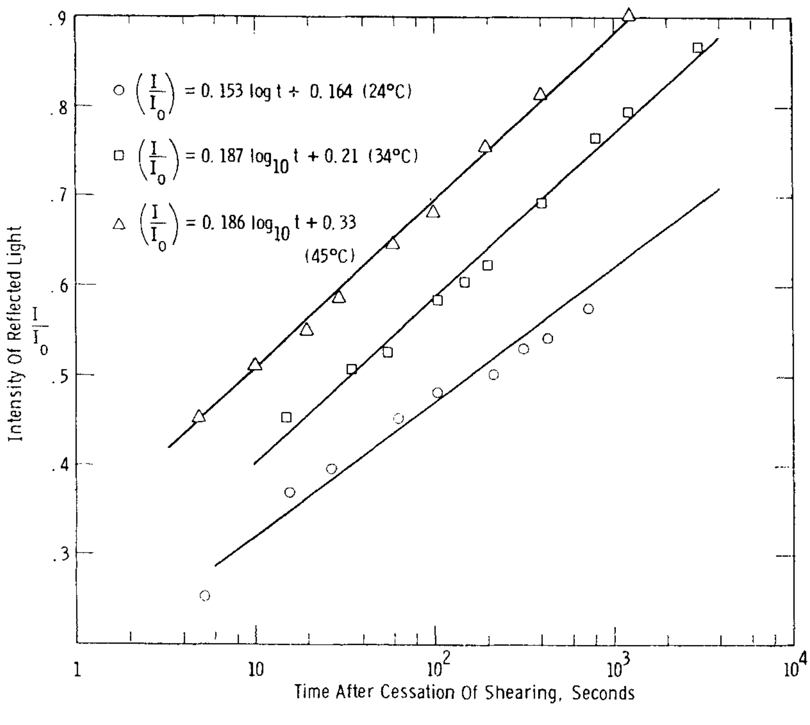

Figure 4. Intensity as a function of relaxation time at 24 °C (○), 34 °C (□), and 45 °C (△).

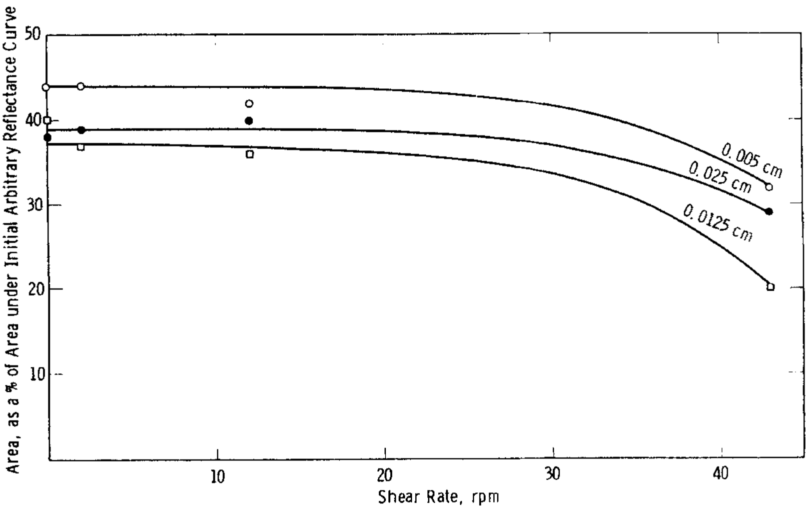

Figure 5. Total reflectance as a function of shear rate for three film thickness.

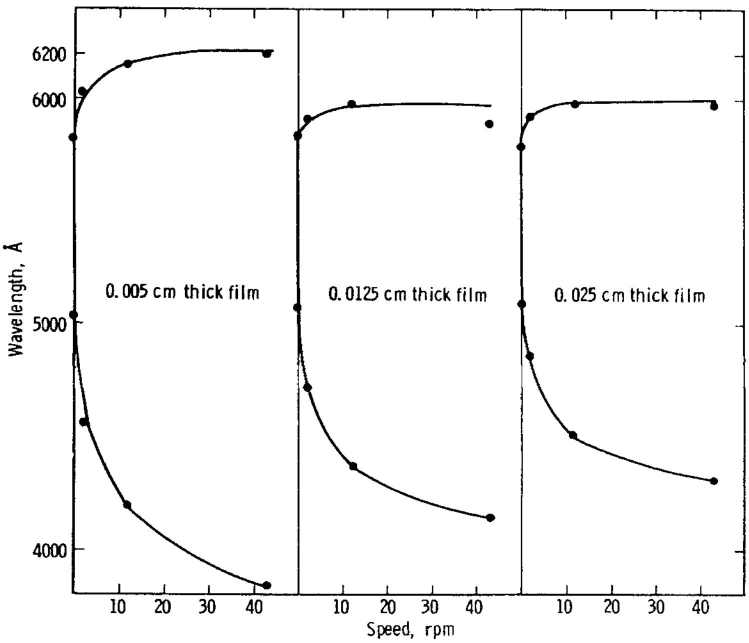

Figure 6. Limits of reflectance as functions of shear rate for three film thicknesses.

Next, the total reflectance was determined as a function of piston velocity, or shear rate, for three liquid crystal film thicknesses (0.025, 0.0125, and 0.005 cm). The behavior of the three films is shown in Fig. 5 where no great difference between the three slopes is observed. The total reflectance drop for all wavelengths is relatively small. However, the spread of the limits of the total reflectance as a function of shear rate does show variations with film thickness: as the thickness of the film increases, the shear rate decreases and reflectance spread decreases, as shown in Fig. 6. Again, it should be noticed that the spread to the lower, blue, end of the spectrum is large relative to the spreading to the higher, red, end.

Finally, the maximum intensity of reflected light is plotted against piston velocity, or shear rate, for three film thicknesses (Fig. 7). The

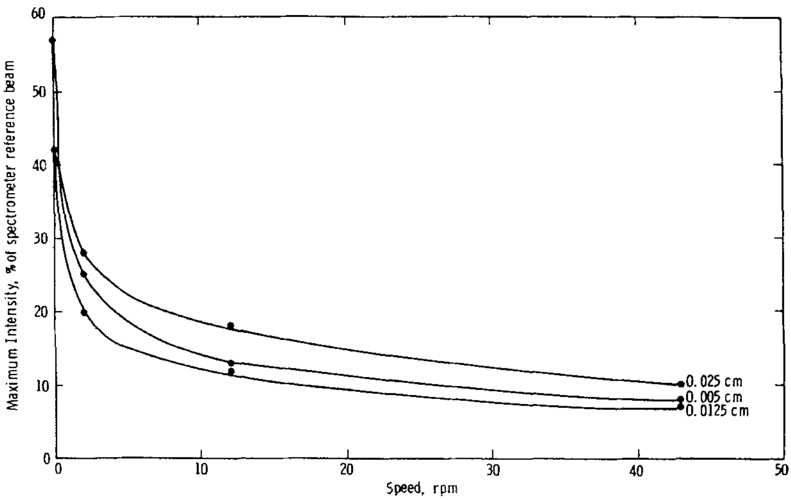

Figure 7. Variation in intensity of peak reflectance wavelength as a function of shear rate for three film thicknesses.

shapes of the three curves are approximately the same, and there does not seem to be any trend regarding the effects of the film thickness other than what might be expected from the higher shear rates for the thin films. It must be remarked here that the drop in intensity as a function of shear rate is considerable.

4. Discussion

On shearing a liquid crystal, one observes a shift of color towards the blue end of the spectrum. However, spectrophotometric results show that this is more apparent than real. Several things are immediately obvious. There is a small shift of peak reflectance wavelength towards the blue (Fig. 2) but, more importantly, there is a decrease in intensity of the peak reflectance wavelength (Figs. 3 and 7) coupled with an increase in the reflecting band width toward the blue end of the spectrum. This is indicated by the dramatic increase in the limits of reflectance (Figs. 3 and 6). However, the total reflectance of the broad band under shear is only slightly less than that of the narrow band under static conditions (Figs. 5 and 6). This was determined by measuring the area under each curve, for runs at various shear rates.

The apparent variation in response with different film thickness (Fig. 6) is due to the fact that any given point in the liquid crystal film is subjected to a lower rate of shear as the film thickness is increased, for a given rate of rotation of the shearing disc, i.e., the thinnest film has the highest gradient and thus shows the greatest effect.

We have used a mechanical model to attempt to explain why the wavelength of maximum reflectance does not change appreciably, while the intensity decreases, on shearing. The system can be approximately described using rectangular coordinates because of the cell geometry, i.e., the cell radius is much greater than the film thickness.† Even for a non-Newtonian fluid, this leads to an expression which shows that there is an approximately linear velocity profile. In this situation, a column of liquid crystal is subjected to drag forces which are proportional to the velocity.

In this model, the liquid crystal helix is bent more and more, on shearing, as it protrudes into the velocity field. The first few periods of the helix (the ones nearer the stationary plate) can remain almost unchanged, as they are at zero velocity; but, as the helix bends more and more, fewer and fewer molecules remain in the position to give

† The motion theory is equivalent to that caused by parallel plates with the top plate moving with a velocity as determined by the radius of the point being observed and the angular velocity of the piston.

the original wavelength of reflected light; therefore, the wavelength remains the same but the intensity decreases. In regions of high velocity, where the drag forces are greater, the van der Waal's forces between liquid crystal molecular layers in the helix are overcome and tumbling may occur. This causes progressive elimination of color response as the tumbling molecules cannot present the correct spatial configuration to the incident light to give reflected light in any wavelength.

This model is illustrated in Figs. 8 and 9. Figure 8 depicts the case

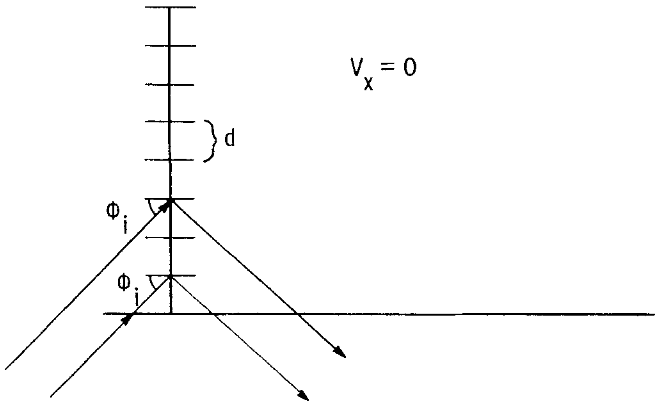

Figure 8. Molecular orientation of liquid crystals at zero shear.

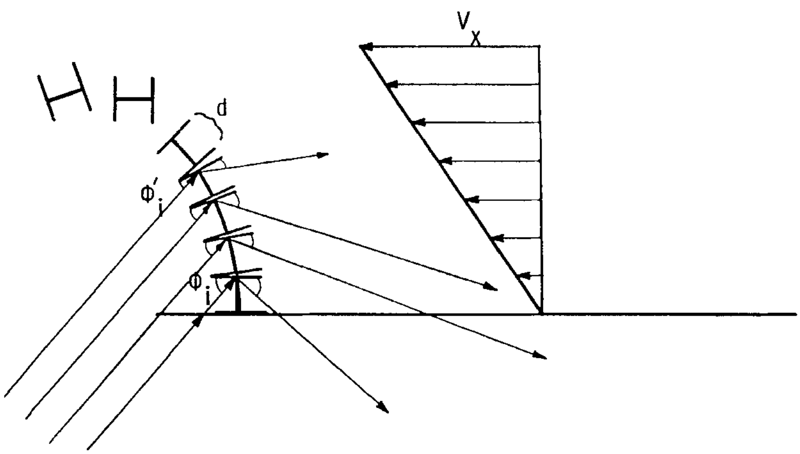

Figure 9. Effects of shear on molecular orientation and consequent light diffraction characteristics.

with zero shear. The layers of liquid crystal have a characteristic spacing d which gives rise to the well-known Bragg scattering, where $\eta\lambda = 2d \sin\phi$ ($\eta = 1, 2, \ldots$). The imposition of a velocity gradient results in the deflection of the column of liquid crystal molecules (Fig. 9).

This mechanism would explain much of the observed data. First, because the velocity approaches zero as one approaches the stationary plate of the apparatus in Fig. 1, the first few layers of liquid crystals are subjected to very small drag forces so they are not disturbed greatly from their positions at rest. Since these layers are at the surface, the contribution to total reflected light due to their orientation is weighted most heavily. This would explain the observed fact that the wavelength of maximum reflectance does not change appreciably on shear.

Layers farther from the stationary surface are deflected more than those close to the stationary surface; this gives rise to a distribution of incident angles ϕ_i, and, consequently, of reflected wavelengths determined by $\eta\lambda_i = 2d \sin\phi_i$. The angle of incidence can be seen to be decreasing; thereby a progression to smaller wavelengths occurs. This accounts for the increasing amount of shorter wavelength light reflected, which is evidenced by the skewing of the reflectance vs wavelength curves to the blue end. Finally, since at distances far from the stationary plate, or at high velocity, the intermolecular forces are overcome, tumbling occurs and the intensity of the reflected light is diminished, as is observed experimentally.

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